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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.	
10/726,744	12/02/2003	Larry C. Olsen	23-65037	6833	
32215	7590 02/29/2008		EXAMINER		
KLARQUIST 1 121 SW SALM	SPARKMAN, LLP 10N STREET, SUITE 1600	BARTON, JEFFREY THOMAS			
ONE WORLD	TRADE CENTER	E CENTER	ART UNIT	PAPER NUMBER	
PORTLAND,	OR 9/204		1795		
			MAIL DATE	DELIVERY MODE	
			02/29/2008	PAPER	

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

	Application No.	Applicant(s)							
	Application No.								
Office Action Summer:	10/726,744	OLSEN ET AL.							
Office Action Summary	Examiner	Art Unit							
	Jeffrey T. Barton	1795							
- The MAILING DATE of this communication app Period for Reply	ears on the cover sneet with the (	correspondence address							
A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTH(S) OR THIRTY (30) DAYS, WHICHEVER IS LONGER, FROM THE MAILING DATE OF THIS COMMUNICATION.  - Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.  - If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.  - Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133).  Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).									
Status									
1) Responsive to communication(s) filed on 29 N	ovember 2007.								
	action is non-final.								
3) Since this application is in condition for allowar closed in accordance with the practice under E									
Disposition of Claims									
4) ⊠ Claim(s) 1-3,5-18,23-25 and 37-39 is/are pend 4a) Of the above claim(s) is/are withdray 5) ☐ Claim(s) is/are allowed. 6) ⊠ Claim(s) 1-3, 5-18, 23-25, and 37-39 is/are rejuictly compared to. 8) ☐ Claim(s) are subject to restriction and/o	wn from consideration.								
Application Papers									
9) The specification is objected to by the Examine 10) The drawing(s) filed on is/are: a) acc		Examiner.							
Applicant may not request that any objection to the	drawing(s) be held in abeyance. Se	ee 37 CFR 1.85(a).							
Replacement drawing sheet(s) including the correct 11) The oath or declaration is objected to by the Ex									
Priority under 35 U.S.C. § 119									
12) Acknowledgment is made of a claim for foreign  a) All b) Some * c) None of:  1. Certified copies of the priority document  2. Certified copies of the priority document  3. Copies of the certified copies of the priority document  application from the International Bureau  * See the attached detailed Office action for a list	s have been received. s have been received in Applica rity documents have been receiv u (PCT Rule 17.2(a)).	tion No red in this National Stage							
Attachment(s)									
1) Notice of References Cited (PTO-892) 2) Notice of Draftsperson's Patent Drawing Review (PTO-948) 3) Information Disclosure Statement(s) (PTO/SB/08) Pager No(s)/Mail Date 20071129	4) Interview Summar Paper No(s)/Mail [ 5) Notice of Informal 6) Other:	Date							

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#### **DETAILED ACTION**

## Response to Amendment

1. The amendment filed on 29 November 2007 does not place the application in condition for allowance.

# Status of Rejections Pending Since the Office Action of 27 June 2007

- 2. All rejections of claims 4 and 19-22 are withdrawn due to cancellation of the claims
- 3. All other rejections are maintained.

# Declaration under 37 C.F.R. §1.131

- 4. The declaration filed on 29 November 2007 under 37 CFR 1.131 has been considered but is ineffective to overcome the Stark et al reference.
- 5. The evidence submitted is insufficient to establish a reduction to practice of the invention in this country or a NAFTA or WTO member country prior to the effective date of the Stark et al reference. The declaration and accompanying exhibit do not demonstrate complete reduction to practice of the instantly claimed invention. Relevant to claims 1 and 37, there is no evidence that a power source comprising the instant electrically conductive member was conceived or reduced to practice prior to the effective filing date of the Stark et al reference. Relevant to claim 23, there is no evidence that a source having a volume less than 10 cm³ and an output from 1 μW to about 1 W was conceived or reduced to practice prior to the effective filing date of Stark

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et al. Also relevant to claim 37, there is no evidence that the claimed coil configuration was conceived or reduced to practice prior to the effective filing date of Stark et al.

In addition, the declaration is only signed by one of the four named inventors in this application. (i.e. Larry C. Olsen) In order to be effective, a declaration under 37 C.F.R. §1.131 must be signed by all named inventors unless it is demonstrated that fewer inventors were responsible for the invention of the subject matter of the claims under rejection. Note MPEP §715.04 as to the formal requirements of declarations under 37 C.F.R. §1.131.

# Claim Rejections - 35 USC § 102

6. The following is a quotation of the appropriate paragraphs of 35 U.S.C. 102 that form the basis for the rejections under this section made in this Office action:

A person shall be entitled to a patent unless -

- (b) the invention was patented or described in a printed publication in this or a foreign country or in public use or on sale in this country, more than one year prior to the date of application for patent in the United States.
- (e) the invention was described in (1) an application for patent, published under section 122(b), by another filed in the United States before the invention by the applicant for patent or (2) a patent granted on an application for patent by another filed in the United States before the invention by the applicant for patent, except that an international application filed under the treaty defined in section 351(a) shall have the effects for purposes of this subsection of an application filed in the United States only if the international application designated the United States and was published under Article 21(2) of such treaty in the English language.
- 7. Claims 1, 5, 6, 13-15, 17-18, and 23 are rejected under 35 U.S.C. 102(b) as being anticipated by DE 297 23 309 U1 (DE '309).

DE '309 discloses a thermoelectric power source as shown in figure 1.

Regarding claims 1 and 5, DE '309 discloses the power source comprising a flexible substrate, 1, having an upper surface and a thermoelectric couple, 4, comprising

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a sputter deposited thin film p-type thermoelement, 4', a sputter deposited thin film ntype thermoelement, 4", and an electrically conductive member, 4", electrically connecting the ends of the thermoelements (figure 2 and page 4, paragraphs 2 to 5). DE '309 teaches bismuth telluride as the thermocouple material. (Page 2, 4<sup>th</sup> paragraph) The stoichiometric formula of bismuth telluride is Bi<sub>2</sub>Te<sub>3</sub>.

Regarding claims 6, 13-15, 17, and 18, the claim limitations; thermoelectric composition, power output, device volume and substrate type, are disclosed within the reference (see page 2 and page 4, paragraphs 2 to 5).

Regarding claim 23, DE '309 discloses the power source comprising a flexible substrate, 1, having an upper surface and a thermoelectric couple, 4, comprising a sputter deposited thin film p-type thermoelement, 4', a sputter deposited thin film n-type thermoelement, 4", and an electrically conductive member, 4", electrically connecting the ends of the thermoelements (figure 2 and page 4, paragraphs 2 to 5). The thermoelectric power source having a volume of less than about 10 cm<sup>3</sup> and a power output from 1 µW to 1 W (page 2, paragraph 4).

Claims 1-3, 5, 10, and 18 are rejected under 35 U.S.C. 102(e) as being 8. anticipated by Stark et al. (U.S.P.G.Pub 2004/0231714).

Stark discloses a thermoelectric power source as shown in figure 1.

Regarding claims 1, 4, and 5, Stark discloses the thermoelectric device is comprised of thin film semiconductors assembled in alternating p- and n-type arrays (figure 2 and paragraph 0029). Stark discloses sputter depositing thin film p-type

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thermoelements, 34, sputter depositing thin film n-type thermoelements, 32, and an electrically conductive metal bridge, 26, connecting the ends of the thermoelements (figure 2 and paragraphs 0032, 0035 and 0044). Stark discloses the use of Bi<sub>2</sub>Te<sub>3</sub>. (Paragraph 0023)

Regarding claims 2 and 3, the dimensions of Stark gives L/A ratios of greater than 100 cm<sup>-1</sup> (thickness of 5 microns, width of 10 microns, length of 100 microns; see paragraphs 0032 and 0034).

Regarding claim 10, figure 3 shows p-type elements having different widths than the n-type elements.

Regarding claim 18, Stark discloses the use of polyimide as the substrate (paragraph 0041).

# Claim Rejections - 35 USC § 103

- 9. The following is a quotation of 35 U.S.C. 103(a) which forms the basis for all obviousness rejections set forth in this Office action:
  - (a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negatived by the manner in which the invention was made.
- 10. The factual inquiries set forth in *Graham* v. *John Deere Co.*, 383 U.S. 1, 148 USPQ 459 (1966), that are applied for establishing a background for determining obviousness under 35 U.S.C. 103(a) are summarized as follows:
  - 1. Determining the scope and contents of the prior art.
  - 2. Ascertaining the differences between the prior art and the claims at issue.
  - 3. Resolving the level of ordinary skill in the pertinent art:

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- 4. Considering objective evidence present in the application indicating obviousness or nonobviousness.
- 11. This application currently names joint inventors. In considering patentability of the claims under 35 U.S.C. 103(a), the examiner presumes that the subject matter of the various claims was commonly owned at the time any inventions covered therein were made absent any evidence to the contrary. Applicant is advised of the obligation under 37 CFR 1.56 to point out the inventor and invention dates of each claim that was not commonly owned at the time a later invention was made in order for the examiner to consider the applicability of 35 U.S.C. 103(c) and potential 35 U.S.C. 102(e), (f) or (g) prior art under 35 U.S.C. 103(a).
- 12. Claims 23-25 are rejected under 35 U.S.C. 103(a) as being unpatentable over Stark et al. (U.S.P.G. Pub 2004/0231714).

Stark discloses a thermoelectric power source as shown in figure 1.

Regarding claim 23, Stark discloses the thermoelectric device is comprised of thin film semiconductors assembled in alternating p- and n-type arrays (figure 2 and paragraph 0029). Stark discloses sputter depositing thin film p-type thermoelements, 34, sputter depositing thin film n-type thermoelements, 32, and an electrically conductive metal bridge, 26, connecting the ends of the thermoelements (figure 2 and paragraphs 0032, 0035 and 0044).

Regarding claim 24, Stark discloses that the thermocouples are connected electrically in series and thermally in parallel, which meets the limitation of the claim. (Figures 1 and 2; Paragraph 0035)

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Regarding claim 25, figure 3 shows p-type elements having different widths than the n-type elements.

The differences between Stark and the claims are the requirements of a specific volume and power output.

The choice of a specific, volume for the device and a power output are dependent on the specific application for the device. Absent any unexpected results, it would have been obvious to one of ordinary skill in the art at the time the invention was made to choose a specific volume and output power as within the claims for the device of Stark. Also Stark discloses the choice of number of thermocouples within the device is dependent on the required power for the device (paragraph 0039), thus making this choice determines the power output and the device size. Therefore the claims are obvious over Stark.

13. Claims 6-9, 11, and 13-16 are rejected under 35 U.S.C. 103(a) as being unpatentable over Stark as applied to claims 1-5, 10, and 18 above.

The disclosure of Stark is as stated above for claims 1-5, 10 and 18. Further, regarding claims 7 and 8, the dimensions disclosed by Stark include at least 0.1 mm in width and at least 20 angstroms in thickness (paragraphs 0032 and 0034). Also Stark discloses the use of greater than 50 or 1000 thermocouples (paragraph 0039).

The differences between Stark and the claims are the requirements of specific power outputs, electrical configurations, volume of the device and element lengths.

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The choice of a specific volume for the device and a power output are dependent on the specific application for the device. The specific wiring methods, series or parallel, also affects the power/current outputs for the device and are well known within the art to alter the wiring to meet the specific requirements of an application. Absent any unexpected results, it would have been obvious to one of ordinary skill in the art at the time the invention was made to choose a specific volume, wiring method and output power as within the claims for the device of Stark. Also Stark discloses the choice of number of thermocouples within the device is dependent on the required power for the device (paragraph 0039), thus making this choice determines the power output and the device size. The choice of element length is a further design choice that is obvious to one skilled in the art. Therefore the claims are obvious over Stark.

14. Claims 12, 17, and 37-39 are rejected under 35 U.S.C. 103(a) as being unpatentable over Stark as applied to claims 1-5, 10 and 18 above, and further in view of Barr et al. (U.S. 4,036,665).

The disclosure of Stark is as stated above for claims 1-5, 10 and 18. The difference between Stark and claims 12 and 37 is the requirement of a coiled substrate configuration. The difference between Stark and claim 17 is the requirement of a specific n-type element. The additional difference between Stark and claim 38 is the required element dimensions. The additional difference between Stark and claim 39 is the required power output.

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Barr teaches a thermopile for a thermoelectric generator. The thermopile is shown in figure 1 and comprises bismuth telluride elements sputter deposited onto a polyimide substrate (abstract). The substrate is coiled up (column 2, lines 58-61). Barr further teaches the use of n-type dopants such as cuprous bromide, silver iodide and antimony iodide (column 2, lines 47-49).

Regarding claims 12 and 37, it would have been obvious to one of ordinary skill in the art at the time the invention was made to coil up the substrate as within Barr for the device of Stark, because the coiled substrate is pencil thin, thus taking up much less space. It would have been further obvious to one of ordinary skill in the art at the time the invention was made to utilize an n-type dopant as within Barr to dope the n-type elements of Stark because Barr discloses dopants are commonly used n-type dopants in the art. Because Barr and Stark are both concerned with thin film thermoelectric devices, one would have a reasonable expectation of success from the combination.

Regarding claims 38 and 39, Stark discloses the choice of number of thermocouples within the device is dependent on the required power for the device (paragraph 0039), thus making this choice determines the power output and the device size. The choice of element length is a further design choice that is obvious to one skilled in the art, absent any evidence of criticality or unexpected results. Therefore the claims are obvious over Stark.

Thus the combination meets the claims.

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# Response to Arguments

15. Applicant's arguments filed 29 November 2007 have been fully considered but they are not persuasive.

Applicant argues that the DE '309 reference does not disclose the claimed formulas, as it teaches only the genus "bismuth telluride". This is not persuasive, because conventional thermoelectric bismuth telluride is Bi<sub>2</sub>Te<sub>3</sub>, as is known by anyone familiar with the thermoelectric art. This formula meets the limitations of the claims. Likewise, this disclosure is enabling, since a skilled artisan reading the reference would have recognized that Bi<sub>2</sub>Te<sub>3</sub> is the material used within DE '309.

Applicant further argues that DE '309 does not teach at least 50 thermoelectric couples as required in claim 6. This limitation is clearly met by the disclosure of the  $2^{nd}$  paragraph of page 2 of the translation. Note that claim 6 does not require the 50 couples to be on a single substrate. Applicant states in describing DE '309 that DE '309 teaches output of 10  $\mu$ W and 3V, which clearly meets the claim limitations.

Applicant further argues that DE '309 does not teach n-type thermoelements that are substantially free of selenium. The Examiner notes that the recited "bismuth telluride" of DE '309 does not include selenium.

Applicant further argues that DE '309 does not disclose a polyimide substrate.

This is incorrect, as polyimide film substrate 1 is clearly disclosed in the 2<sup>nd</sup> paragraph of page 4 of the translation. A polyimide film substrate is flexible.

Applicant's contention that DE '309 teaches away from rolling a flexible substrate is not relevant to any of the claims that are rejected as anticipated by this reference.

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Applicant's arguments that Stark is not available as prior art are not persuasive due to the deficiencies in the declaration under 37 C.F.R. §1.131, as noted above.

#### Conclusion

16. Applicant's amendment necessitated the new ground(s) of rejection presented in this Office action. Accordingly, **THIS ACTION IS MADE FINAL**. See MPEP § 706.07(a). Applicant is reminded of the extension of time policy as set forth in 37 CFR 1.136(a).

A shortened statutory period for reply to this final action is set to expire THREE MONTHS from the mailing date of this action. In the event a first reply is filed within TWO MONTHS of the mailing date of this final action and the advisory action is not mailed until after the end of the THREE-MONTH shortened statutory period, then the shortened statutory period will expire on the date the advisory action is mailed, and any extension fee pursuant to 37 CFR 1.136(a) will be calculated from the mailing date of the advisory action. In no event, however, will the statutory period for reply expire later than SIX MONTHS from the date of this final action.

17. Any inquiry concerning this communication or earlier communications from the examiner should be directed to Dr. Jeffrey T. Barton whose telephone number is (571)272-1307. The examiner can normally be reached on M-F 9:00AM - 5:30PM.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Nam Nguyen can be reached on (571) 272-1342. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

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Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a USPTO Customer Service Representative or access to the automated information system, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

/Nam X Nguyen/ Supervisory Patent Examiner, Art Unit 1753

JTB 13 February 2008

# PATENT COOPERATION TREATY

# **PCT**

# INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference 23 - 65037 - 03	FOR FURTHER ACTION	see Form PCT/ISA/220 as well as, where applicable, item 5 below.
International application No.	International filing date (day/mont	h/year) (Earliest) Priority Date (day/month/year)
PCT/US2004/040460	02/12/2004	02/12/2003
Applicant		
BATTELLE MEMORIAL INSTITU	TE	
This International Search Report has bee according to Article 18. A copy is being tr		rching Authority and is transmitted to the applicant u.
This International Search Report consists  X  It is also accompanied by	of a total of8short a copy of each prior art document of	
	international search was carried out less otherwise indicated under this it	on the basis of the international application in the lem.
The international this Authority (Ru		of a translation of the international application furnished to
b. With regard to any nucle	otide and/or amino acid sequence	e disclosed in the international application, see Box No. I.
2. Certain claims were fou	ind unsearchable (See Box II).	
3. X Unity of invention is lac	king (see Box III).	
4. With regard to the title,		
the text is approved as su	ubmitted by the applicant. shed by this Authority to read as follo	we.
L	since by this nationly to read as ising	
5. With regard to the abstract,		
X the text is approved as su	ubmitted by the applicant.	
the text has been establis may, within one month fro	shed, according to Rule 38.2(b), by to om the date of mailing of this interna	his Authority as it appears in Box No. IV. The applicant tional search report, submit comments to this Authority.
6. With regard to the drawings,		
a. the figure of the drawings to be p		No. <u>15</u>
X as suggested by	the applicant.  is Authority, because the applicant fa	ailed to suggest a figure
<u> </u>	is Authority, because this figure bette	•
b. none of the figures is to b	e published with the abstract.	

International application No. PCT/US2004/040460

# INTERNATIONAL SEARCH REPORT

Box II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)
This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:
1. Claims Nos.: because they relate to subject matter not required to be searched by this Authority, namely:
Claims Nos.:     because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:
Claims Nos.:     because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).
Box III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)
This International Searching Authority found multiple inventions in this international application, as follows:
see additional sheet
As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.
2. As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:
4. No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:
Remark on Protest  The additional search fees were accompanied by the applicant's protest.  X  No protest accompanied the payment of additional search fees.

# FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. claims: 1-36

A thermoelectric power source comprising a flexible substrate and a sputtered thin film of thermoelectric material and method of making it.

2. claims: 37-66

A method and apparatus for providing electrical energy by transmitting ambient energy to a thermoelectric device

3. claims: 67-85

A thermoelectric power source comprising a sputtered film of thermoelectric material and a high temperature heat pipe and a low temperature heat pipe; and/or a thermoelectric power source comprising a thin film TE module formed on a flexible substrate and a reel about which the flexible substrate is wound.

International Application No
PCT/US2004/040460

A CLASSIFICATION OF SUBJECT MATTER
H01L35/30 H01L H01L35/08 According to International Patent Classification (IPC) or to both national classification and IPC Minimum documentation searched (classification system followed by classification symbols) H011 Documentation searched other than minimum documentation to the extent that such documents are included, in the fields searched Electronic data base consulted during the international search (name of data base and, where practical, search terms used) EPO-Internal, PAJ, INSPEC, WPI Data C. DOCUMENTS CONSIDERED TO BE RELEVANT Category " Citation of document, with indication, where appropriate, of the relevant passages Relevant to claim No. χ DE 297 23 309 UI (D.T.S. GESELLSCHAFT ZUR 1,4-6, FERTIGUNG VON 13-15, DUENNSCHICHT-THERMOGENERATOR-SY) 17,18, 10 September 1998 (1998-09-10) 23,24, 26,27, 30,32,33 Υ page 4, paragraph 2 - paragraph 5; figures 67 - 75Х US 4 036 665 A (BARR ET AL) 1,4-6,19 July 1977 (1977-07-19) 18,26, 30,31,33 column 2, line 30 - column 4, line 33; figures 1,2Α 34-36 -/--Further documents are listed in the continuation of box C. Patent family members are listed in annex " Special categories of cited documents : "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory, underlying the "A" document defining the general state of the lart which is not considered to be of particular relevance invention "E" earlier document but published on or after the international "X" document of particular relevance; the claimed invention filing date cannot be considered novel or cannot be considered, to document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention citation or other special reason (as specified) cannot be considered to involve an inventive step when the document is combined with one or more other, such docu-"O" document referring to an oral disclosure, use, exhibition or ments, such combination being obvious to a person skilled in the art. other means document published prior to the international filing date but later than the priority date claimed "8" document member of the same patent family Date of the actual completion of the international search Date of mailing of the international search report N 7 MAARI 2006 28 February 2006 Name and mailing address of the ISA Authorized officer European Patent Office, P.B. 5818 Patentläan 2 NL – 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo ni, Kirkwood, J Fax: (+31-70) 340-3016

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International Application No PCT/US2004/040460

Category '	Cration of decument, with indication, where appropriate, of the relicant passages	Relevant to claim No
X A	US 3 554 815 A (ROBERT OTTO OSBORN) 12 January 1971 (1971-01-12) the whole document	1,4,5,8, 12,18, 26,30, 31,76, 80,82-85 2,3,28, 29,67, 70,71
Р,Х	US 2004/231714 A1 (STARK INGO ET AL) 25 November 2004 (2004-11-25)	1-18, 23-30, 32-36
E	the whole document -& WO 2004/105143 A (APPLIED DIGITAL SOLUTIONS; STARK, INGO; ZHOU, PETER) 2 December 2004 (2004-12-02)	1-18, 23-30, 32-36
A	WO 03/007391 A (FERROTEC CORPORATION) 23 January 2003 (2003-01-23) the whole document	1-36
Α	US 5 286 304 A (MACRIS ET AL) 15 February 1994 (1994-02-15) the whole document	1-36,67, 70,71,76
A Y	WO 02/23642 A (FRAUNHOFER-GESELLSCHAFT ZUR FOERDERUNG DER ANGEWANDTEN FORSCHUNG E.V;) 2] March 2002 (2002-03-21) figures 11a-18c	1-36,67, 70,71,76 67-75
A	PATENT ABSTRACTS OF JAPAN vol. 1997, no. 08, 29 August 1997 (1997-08-29) & JP 09 107129 A (SHARP CORP), 22 April 1997 (1997-04-22) abstract	1-36,67, 70,71,76
X	SCHMIDT F: "Batterielose Funksensoren, betrieben mit Energie aus der Umgebung" 11 March 2002 (2002-03-11), XP002231431 Retrieved from the Internet: URL:http://www.enocean.com/comments/tagung 0302.pdf> 'retrieved on 2006-02-28! the whole document	37-66
Х	WO 03/015186 A (ENOCEAN GMBH; ALBSMEIER, ANDRE; BULST, WOLF-ECKARDT; PISTOR, KLAUS; SC) 20 February 2003 (2003-02-20) the whole document	37-66
X	WO 02/095707 A (ENOCEAN GMBH; ALBSMEIER, ANDRE; BULST, WOLF-ECKHART; PISTOR, KLAUS; SC) 28 November 2002 (2002-11-28) the whole document	37-66

International Application No PCT/US2004/040460

<del></del>	ilion) DOCUMENTS CONSIDERED TO BE RELEVANT	
Category "	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No
A	US 6 232 543 B1 (NAGATA YOICHI) 15 May 2001 (2001-05-15) the whole document	37-66
Y	PATENT ABSTRACTS OF JAPAN vol. 1997, no. 12, 25 December 1997 (1997-12-25) -& JP 09 224387 A (WAKABAYASHI TADATOSHI), 26 August 1997 (1997-08-26) abstract	67-85
Y	PATENT ABSTRACTS OF JAPAN vol. 2003, no. 10, 8 October 2003 (2003-10-08) -& JP 2003 179275 A (YASKAWA ELECTRIC CORP), 27 June 2003 (2003-06-27) abstract; figure 6	67-85
А	US 4 520 305 A (CAUCHY ET AL) 28 May 1985 (1985-05-28) figures 9,11	67-85
Α	US 4 312 402 A (BASIULIS ET AL) 26 January 1982 (1982-01-26) the whole document	67-85
A .	US 2002/148236 A1 (BELL LON E) 17 October 2002 (2002-10-17) the whole document	67-75
A	US 2002/139123 A1 (BELL LON E) 3 October 2002 (2002-10-03) the whole document	67-75

Information on patent family members

PCT/US2004/040460

	atent document d in search report		Publication date		Patent lamity member(s)		Publication date
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# PATENT COOPERATION TREATY

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To: see form PCT/ISA/220				PCT  WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORIT  (PCT Rule 43bis.1)		
Applicant's or ag				FOR FURTHI See paragraph 2		
International app PCT/US2004			International filing date 02.12.2004	(day/month/year)	Priority date (day/month/year) 02.12.2003	
International Pat H01L35/30, H			both national classification	n and IPC DOCK	ETED FOR: 4/7/04	
Applicant BATTELLE N	иЕМОР	RIAL INSTITU	TE		UTER MA	
1. This opi	nion co	ontains indicat	ions relating to the fo	SCAN following items.		
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☐ Box N		-	ment of opinion with red	nard to novelty, inv	entive step and industrial applicability	
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⊠ Box N		Reasoned sta			d to novelty, inventive step or industrial statement	
⊠ Box N	No. VI	Certain docun	nents cited			
☐ Box N	No. VII	Certain defect	s in the international ap	plication		
⊠ Box N	No. VIII	Certain obser	vations on the internation	onal application		
2. FURTHE	R ACT	ION				
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# WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY

International application No. PCT/US2004/040460

	Вох	No. I Basis of the opinion							
1.	<ol> <li>With regard to the language, this opinion has been established on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.</li> </ol>								
	This opinion has been established on the basis of a translation from the original language into the following language, which is the language of a translation furnished for the purposes of international search (under Rules 12.3 and 23.1(b)).								
2.	2. With regard to any nucleotide and/or amino acid sequence disclosed in the international application and necessary to the claimed invention, this opinion has been established on the basis of:								
	a. typ	pe of material:							
		a sequence listing							
		table(s) related to the sequence listing							
b. format of material:									
		in written format							
		in computer readable form							
	c. tim	ne of filing/furnishing:							
		contained in the international application as filed.							
		filed together with the international application in computer readable form.							
		furnished subsequently to this Authority for the purposes of search.							
3.	1	n addition, in the case that more than one version or copy of a sequence listing and/or table relating thereto has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that in the application as filed or does not go beyond the application as filed, as appropriate, were furnished.							
4.	Addit	ional comments:							

# WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY

International application No. PCT/US2004/040460

	Bo	x No. IV	Lack of unity of	inventior	1					
1.	Ø	☐ In response to the invitation (Form PCT/ISA/206) to pay additional fees, the applicant has:								
		⊠	paid additional fees	S.						
			paid additional fees	under pr	otest.					
			not paid additional	fees.						
2.		This A the app	uthority found that th plicant to pay additio	e require nal fees.	ment of ur	nity of invention is not complied with and chose not to invite				
3.	Thi	s Autho	rity considers that th	e requirer	ment of un	ity of invention in accordance with Rule 13.1, 13.2 and 13.3 is				
		complie	d with							
	$\boxtimes$	not com	plied with for the foll	owing rea	asons:					
		see se	eparate sheet							
4.	Co	nsequer	ntly, this report has b	een estat	olished in r	respect of the following parts of the international application:				
	⋈	☑ all parts.								
		the part	s relating to claims N	los						
		ine part	o relating to elatine r	100.						
	Bo	x No. V	Reasoned state	ment und	ler Bule 4:	3bis.1(a)(i) with regard to novelty, inventive step or				
_			applicability; citation	ons and e	explanatio	ons supporting such statement				
1.	Sta	itement								
	No	velty (N)	)	Yes: No:	Claims Claims	2,3,7-12, 16, 19-22, 28, 29, 31, 34-36,67-85 1, 4-6, 13-15, 17-18, 23,27,30,32,33,37-66				
	Inv	entive s	tep (IS)	Yes: No:	Claims Claims	1-85				
	Ind	ustrial a	applicability (IA)	Yes: No:	Claims Claims	1-85				
2	Cit	ations a	nd explanations							

see separate sheet

# WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY

International application No. PCT/US2004/040460

#### Box No. VI Certain documents cited

- Certain published documents (Rules 43bis.1 and 70.10) and /or
- 2. Non-written disclosures (Rules 43bis.1 and 70.9)

see form 210

# Box No. VIII Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet

Reference is made to the following documents:

- D1: DE 297 23 309 U1 (D.T.S. GESELLSCHAFT ZUR FERTIGUNG VON DUENNSCHICHT-THERMOGENERATOR-SY) 10 September 1998 (1998-09-10)
- D2: WO 2004/105143 (APPLIED DIGITAL SOLUTIONS) 2 December 2004 (02.12.04)
- D3: PATENT ABSTRACTS OF JAPAN vol. 1997, no. 08, 29 August 1997 (1997-08-29) & JP 09 107129 A (SHARP CORP), 22 April 1997 (1997-04-22)
- D4: SCHMIDT F: "Batterielose Funksensoren, betrieben mit Energie aus der Umgebung" 11 March 2002 (2002-03-11), XP002231431 Retrieved from the Internet:
- URL:http://www.enocean.com/comments/tagung 0302.pdf> [retrieved on 2006-02-28]
- D5: PATENT ABSTRACTS OF JAPAN vol. 1997, no. 12, 25 December 1997 (1997-12-
- 25) -& JP 09 224387 A (WAKABAYASHI TADATOSHI), 26 August 1997 (1997-08-26)
- D6: US-A-3 554 815 (ROBERT OTTO OSBORN) 12 January 1971 (1971-01-12)
- D7: WO 02/23642 A (FRAUNHOFER-GESELLSCHAFT ZUR FOERDERUNG DER ANGEWANDTEN FORSCHUNG E.V;) 21 March 2002 (2002-03-21)
- D8: PATENT ABSTRACTS OF JAPAN vol. 2003, no. 10, 8 October 2003 (2003-10-08) & JP 2003 179275 A (YASKAWA ELECTRIC CORP), 27 June 2003 (2003-06-27)

# Re Item IV

#### Lack of unity of invention

This Authority considers that there are three inventions covered by the claims indicated as follows:

#### 1. Claims 1-36

A thermoelectric power source comprising a flexible substrate and a sputtered thin film of thermoelectric material and method of making it.

#### 2. Claims 37-66

A method and apparatus for providing electrical energy by transmitting ambient energy to a thermoelectric device

#### 3. Claims 67-85

A thermoelectric power source comprising a sputtered film of thermoelectric material and a high temperature heat pipe and a low temperature heat pipe; and/or a thermoelectric

# WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY (SEPARATE SHEET)

power source comprising a thin film TE module formed on a flexible substrate and a reel about which the flexible substrate is wound.

The reasons for which the inventions are not so linked as to form a single general inventive concept, as required by Rule 13.1 PCT, are as follows:

Rule 13.1 PCT requires that the international application shall relate to one invention only or to a group of inventions so linked as to form a single general inventive concept ("requirement of unity of invention"). According to Rule 13.2 PCT, where a group of inventions is claimed in one and the same international application, the requirement of unity of invention referred to in Rule 13.1 shall be fulfilled only when there is a technical relationship among those inventions involving one or more of the same or corresponding special technical features. The expression "special technical features" shall mean those technical features that define a contribution which each of the claimed inventions, considered as a whole, makes over the prior art.

The prior art has been identified as document DE 297 23 309 U which discloses a thermoelectric power source comprising a flexible substrate and a thermoelectric couple comprising sputter deposited thin films. Therefore claim 1 contains no technical feature which makes a contribution over the prior art and can be considered as a special technical feature within the meaning of Rule 13.2 PCT. Therefore the first invention (claims 1-36) cannot be linked to any other invention by special technical features.

Claims 37-66 relate to a method and apparatus for providing electrical energy by transmitting ambient energy to a thermoelectric device. A priori there are no special technical features in common with the first or third named invention.

Furthermore claims 67-85 relate to a thermoelectric power source comprising a sputtered film of thermoelectric material and a high temperature heat pipe and a low temperature heat pipe; and/or a thermoelectric power source comprising a thin film TE module formed on a flexible substrate and a reel about which the flexible substrate is wound. This invention has matter in common with the first invention of "a thermoelectric power source comprising a sputtered film of thermoelectric material". However, as this matter is not new in view of the document named above, there are no common special technical features.

In conclusion, the groups of claims are not linked by common or corresponding special technical features and define three different inventions not linked by a single general inventive concept.

The application, hence does not meet the requirements of unity of invention as defined in Rules 13.1 and 13.2 PCT.

#### Re Item V

Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

### Invention 1, Claims 1-36

The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 1, 4-6, 13-15, 17-18, 23, 27, 30, 32,33 is not new in the sense of Article 33(2) PCT and because the subject-matter of claims 2,3,7-12, 16, 19-22, 28, 29, 31, 34-36 does not involve an inventive step in the sense of Article 33(3) PCT.

#### 1.1 INDEPENDENT CLAIM 1

The document D1 discloses (the references in parentheses applying to this document, cf. Figs 2-5 and page 4, pgh 2 to 5):

- a thermoelectric power source comprising:
- a flexible substrate (polyimide film 1) having an upper surface; and
- a thermoelectric couple (4) comprising:
- (a) a sputter deposited thin film p-type thermoelement (4') positioned on the upper surface of the flexible substrate:
- (b) a sputter deposited thin film n-type thermoelement (4") positioned on the upper surface of the flexible substrate adjacent the p-type thermoelement; and
- (c) an electrically conductive member (Metallbrücke 4") positioned on the flexible substrate and electrically connecting a first end of the p-type thermo element with a second end of the n-type thermoelement.
- 1.2 The additional features of claims 4-6, 13-15 and 17-18 are also known from D1, see passages cited above.

The additional features of claims 2, 3 when interpreted from the description as length of leg of the thermoelement divided by the cross sectional area are obvious to a skilled person in view of the Figures. Since the thermoelements of D1 are made of a thin film material, the cross sectional area will be significantly smaller than the length. This is considered as falling within the limits given in these claims.

Likewise the specific lengths and thicknesses given in claims 7 and 8 are considered as falling within the range expected by a skilled person reading D1.

The additional features of claim 9 regarding the power output would be regarded by the skilled person as achievable using the device of D1 with a sufficient number of thermocouples.

The geometric arrangement cited in claim 10 is known from document D2, which is a prior filed document but published on the filing date of this application.

The circuit arrangements according to claims 11 and 16 are obvious variations for a skilled person, for example to reduce overall resistance of the device.

The additional feature of claim 12 of the flexible substrate in a coil configuration is known from document D3, see abstract and is an obvious alternative to the stacked arrangement shown in D1.

The use of a superlattice thermoelement on a thin film device according to claims 19-22 is already known from document D3.

#### 1.3 INDEPENDENT CLAIM 23

The document D1 discloses (the references in parentheses applying to this document, cf. Figs 2-5 and page 4, pgh 2 to 5):

- a thermoelectric power source comprising:
- a flexible substrate (polyimide film 1) having an upper surface;
- multiple thermocouples (4) electrically connected to one another on the upper surface of

the flexible substrate, the thermocouples comprising:

sputter deposited thin film p-type thermoelements (4'):

sputter deposited thin film n-type thermoelements (4") alternatingly positioned adjacent the p-type thermoelements; and

wherein the thermoelectric power source has a volume of less than about 10 cm3 and has a power output of from about 1 microWatt to about 1 W (see page 4, pgh 4).

Therefore the subject matter of claim 23 is not new.

The additional features of claims 24 and 25 are obvious to a skilled person, see reasoning above for claims 10 and 11.

# 1.4 INDEPENDENT CLAIM 26

The document D1 discloses (the references in parentheses applying to this document, cf. Figs 2-5 and page 4, pgh 2 to 5):

a method for fabricating thermoelectric power sources comprising:

providing a flexible substrate (polyimide film 1);

sputter depositing multiple thin films of n-type thermoelectric material (4') onto the flexible substrate:

sputter depositing multiple thin films of p-type thermoelectric material (4") onto the flexible substrate; and

forming multiple thermocouples (4) on the flexible substrate by electrically connecting (Metallbrücke 4"") the thin films of p-type thermoelectric material to the thin films of n-type thermoelectric materials.

Therefore the subject matter of claim 26 is not new.

The additional features of claims 27, 30, 32 and 33 are known from D1, see passage referenced above.

The additional features of claims 28 and 29 are obvious to a skilled person in view of D1. The additional feature of claim 31 of further comprising winding the flexible substrate into a coil configuration is known from document D3 and is an obvious alternative to the stacked

arrangement shown in D1.

The additional features of claims 34-36 concern standard conditions for sputtering which would be selected routinely by a person skilled in the art.

#### 2. Invention 2, Claims 37-66

The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 37-49 is not new in the sense of Article 33(2) PCT.

#### 2.1 INDEPENDENT CLAIM 37

The document D4 discloses (the references in parentheses applying to this document, cf. Bild 1, 7 and 8):

A method for providing electrical energy to an electrical device (thermally powered transmitter) in an environment having a first and a second temperature region comprising the steps of:

providing a means for transmitting ambient energy collected in the first temperature region (Umgebungsenergie),

providing a thermoelectric device having a first side and a second side (Energiewandler), providing the means for transmitting the ambient energy collected in the first temperature region in communication with the first side of the thermoelectric device (contact by conduction), and

providing the second side of the thermoelectric device in communication with the second temperature region.

- 2.2 Dependent claims 38-49 do not contain any features which, in combination with the features of any claim to which they refer, meet the requirements of the PCT in respect of novelty and/or inventive step, see D4.
- 2.3 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 50-66 is not new in the sense of Article 33(2) PCT.

**INDEPENDENT CLAIM 50** 

The document D4 discloses (the references in parentheses applying to this document, cf. Bild 1, 7 and 8) an apparatus for generating electrical energy from an environment (Umgebungsenergie) having a first temperature region and a second temperature region comprising a thermoelectric device (Energiewandler) having a first side and a second side wherein the first side is in communication with a means for transmitting ambient thermal energy collected in the first temperature region.

2.4 Dependent claims 51-66 do not contain any features which, in combination with the features of any claim to which they refer, meet the requirements of the PCT in respect of novelty and/or inventive step, see D4.

# 3. Invention 3, Claims 67-85

3.1 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 67-75 does not involve an inventive step in the sense of Article 33(3) PCT.

#### **INDEPENDENT CLAIM 67**

The document D5 discloses (the references in parentheses applying to this document, cf. Figs 5 and 6):

A thermoelectric power source comprising:

- a thermoelectric module (1) having at least one thermoelectric couple comprising a p-type thermoelement (2), an n-type thermoelement (3) positioned adjacent the p-type thermoelement, and an electrically conductive member (electrode 4,5) electrically connecting a first end of the p-type thermoelement with a second end of the n-type thermoelement;
- a high-temperature heat pipe (22a) connected to a hot connection of the thermoelectric module; and
- a low-temperature heat pipe (22b) connected to a cold connection of the thermoelectric module.

from which the subject matter of claim 67 differs in that the thermoelectric element is a sputter deposited thin film.

The problem to be solved may be regarded as seeking a more easily mass produced thermoelectric module.

The solution is obvious to a skilled person since it is known from Document D1 (Fig 2) or D7 (see Fig 17a) to make thermoelectric modules from sputter deposited thin film thermoelectric material.

3.2 Dependent claims 68-75 do not contain any features which, in combination with the features of any claim to which they refer, meet the requirements of the PCT in respect of novelty and/or inventive step, see D6, D1 and D7.

#### 3.3 INDEPENDENT CLAIM 76

The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 76-85 does not involve an inventive step in the sense of Article 33(3) PCT.

The document D6 discloses (the references in parentheses applying to this document):

- a TE device (cf. Fig. 6) comprising:
- a thin film TE module comprising multiple thin film TE p-type and n-type elements formed on a flexible substrate (37);
- a reel having a first end and a second end and about which the flexible substrate is wound (implicit from diagram, fins 45 serves as core of the reel);
- a low-temperature member thermally connected to the first end of the reel (heat exchanger 42); and
- a high-temperature (member) connected to the second end of the reel (heat exchanger 43), wherein the low-temperature and high-temperature members transfer heat to the and from the TE module.

from which the subject matter of claim 76 differs in that the TE module is a power source whereas in D6 power is supplied to the TE device (i.e. it is in cooling mode).

This difference is immaterial to the construction of the claimed device since a skilled person knows that if a temperature difference is established between the different heat

exchangers, a voltage will be generated by virtue of the Seebeck effect.

The same considerations apply to the device of document D5 in combination with D1 or D8

3.4 Dependent claims 77-85 do not contain any features which, in combination with the features of any claim to which they refer, meet the requirements of the PCT in respect of inventive step.

For claims 77-79, 81 refer to document D5. For claims 80-85 refer to D6.

#### Re Item VI

#### Certain documents cited

#### Certain published documents

Application No	Publication date (day/month/year)	Filing date	Priority date (valid claim)
Patent No		(day/month/year)	(day/month/year)
WO2004/105143	02.12.2004	13.05.2004	19.05.2003

# Re Item VIII

#### Certain observations on the international application

The application does not meet the requirements of Article 6 PCT, because claims 2, 3, 28 and 29 are not clear.

Claims 2, 3, 28 and 29: The term "L/A ratios" is not defined with respect to any other parameters in the claim, rendering them unclear. However this term has been interpreted using the description for the purposes of this report.

Although claims 1 and 23 have been drafted as separate independent claims, they appear to relate effectively to the same subject-matter (thermoelectric power source) and to differ from each other only with regard to the definition of the subject-matter for which protection is sought and/or in respect of the terminology used for the features of that subject-matter. The aforementioned claims therefore lack conciseness and as such do not meet the requirements of Article 6 PCT.

# Optimisation of p - $(Bi_{0.25}Sb_{0.75})_2Te_3$ and n - $Bi_2(Te_{0.9}Se_{0.1})_3$ Films for Thermoelectric Thin Film Components

M. STÖLZER, M. STORDEUR, Martin-Luther-Univ. Halle, FB Physlk, D-06099 Halle, Tel/Fax: (0345) 5511100 I. STARK, tecom Bernd & Schöpke ThermoElektrische Componenten, Schwoitscher Str. 2, D-06184 Gröbers

#### Introduction

The trigonal  $V_2VI_3$  compound semiconductors are the most common materials for thermoelectric applications working near room temperature because of their high thermoelectric figure of merit Z.

The aim of our work is to develop films of those materials that could be used in miniaturized thermoelectric coolers or generators in the power range < 1 mW. For these applications, films of several  $\mu m$  (1 . . . 100) thickness and best available Z values are needed.

The figure of merit Z depends on the carrier mobility  $\mu$  and on the carrier density (via the reduced fermi level  $\eta$ ) nearly like

$$Z = \frac{S^2 \sigma}{\kappa} \propto \mu m_d^{3/2} \frac{1}{\kappa_L} f(\eta)$$

 $(m_d$ : density of states mass,  $\kappa_L$ : lattice thermal conductivity). Consequently, best Z values are reached at high mobilities (single crystal-like as possible) and optimized carrier densities (in actual practice some  $10^{19}$  cm<sup>-3</sup>).

### Film deposition

Films of 1 . . . 5 µm thickness were deposited on  $Al/Al_2O_3$ ,  $Si/Si_2O_3$  and Kapton (polyimid) substrates by means of a flash evaporation technique (see fig. 1) with deposition rates of about 1 nm/s. The substrate temperature was varied between 80 °C and 300°C, the optimum was found around 220°C.

Kapton foil is of special interest because of its thermal expansion coefficient  $\alpha=2*10^{-5}$  is nearly the same as that of  $Bi_2Te_3$  and  $Sb_2Te_3$ .

To reach stoichiometric composition of the films the evaporant consists of either  $(Bi_{0.25}Sb_{0.75})_2Te_3$  or  $Bi_2(Te_{0.9}Se_{0.1})_3 + 0.04$  J and some at% additional Tellurium .The

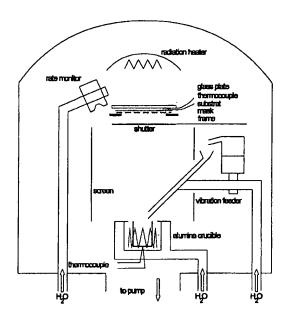


Fig. 1: Schematic of flash evaporation apparatus

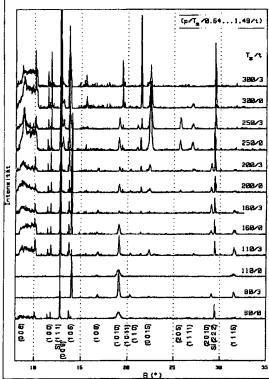


Fig. 2: XRD pattern of (Bi<sub>0.25</sub>Sb<sub>0.75</sub>)<sub>2</sub>Te<sub>3</sub> films T<sub>s</sub> - substrate temperature, t - annealing time

material was homogenized at 850 °C and milled to a grain size of about 100 µm in diameter. After deposition, the films were annealed for 1 . . . 8 hours at 300°C in small evacuated ampoules.

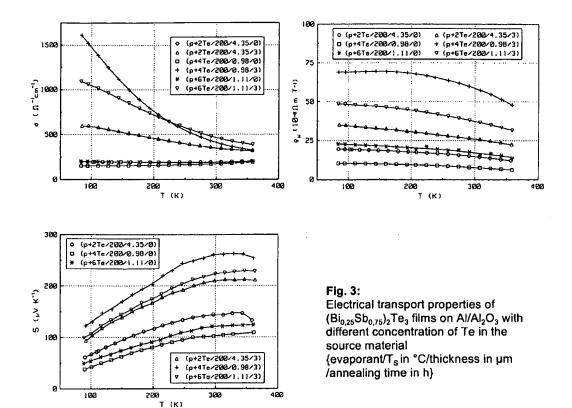
# Optimization of thermoelectric properties.

The prepared films are polycrystalline (except at T<sub>s</sub><100 °C) and show some texture, depending on substra te temperature, film composition and substrate material.

The XRD pattern in fig. 2 illustrate the typical depending on  $T_{\rm s}$  of the texture. At 80 and 110 °C (as grown films) there are only weak and broad peaks because the films are con sisted of very small grains or they are partially amorphous. Annealed films and those grown at higher  $T_{\rm s}$  up to 200 °C show some preferential orientation in the (1 0 5) direction.

At  $T_s$ = 250 °C a strong (0 0 n) texture occurs (c-axis perpendicular to the substrate surface) which is seen by the (0 0 6), (0 0 9) and (0 0 15) peaks. At even higher  $T_s$  there are grains of different orientations growing.

From the point of view of application, the (0 0 n) texture is preferred because of the highly anisotropic electrical conductivity in (Bi<sub>0.25</sub>Sb<sub>0.75</sub>)<sub>2</sub>Te<sub>3</sub> single crystals having its largest value



perpendicular to the (0 0 n) (i. e. trigonal) direction. However, the electrical conductivity of annealed films grown at 200 and 250 °C is nearly the same.

Fig. 3 shows the electrical conductivity, the Hall and the Seebeck effect vs. tempe rature of as grown and annealed  $(Bi_{0.25}Sb_{0.75})_2Te_3$  films. The pictures show the optimisation of thermoelectric properties by variation of the contents of Te in the source material. After the films had been annealed (3 h 300°C), the electrical conductivity is grown up dramatically due to the grown carrier mobility. The hall and the seebeck coefficients had been increased, too, showing a decreased carrier concentration.

The greatest influence of the annealing procedure was reached at 4 at% additional Te in the evaporant, where the mobility at room temperature reaches more then 70% of the value in a single crystal.

Fig. 4 shows the thermoelectric power factor  $S^2\sigma$ , the hall mobility  $\mu_H$  and the carrier concentration p of annealed  $(Bi_{0.25}Sb_{0.75})_2Te_3$  films of different thickness and for a comparison of a single crystal with high Te concentration.

Films on Kapton reach even higher mobilities as on Al<sub>2</sub>O<sub>3</sub>, probably because of the better thermal expansion behaviour of the Kapton substrate. However, if the thickness of the films is greater, the mobility becomes smaller and the carrier

Properties of (Bi<sub>0.25</sub> Sb<sub>0.75</sub>)<sub>2</sub>Te<sub>3</sub> films grown at T<sub>s</sub>=200...230°C by flash evaporation Source material:  $(Bi_{0.25} Sb_{0.75})_2 Te_3 + add$ . Te 5°d/ 10-3 WK-2 m.1  $d(\mu m)$ substrat A+A2O3 0.98 1.0 Kapton Kapton 1.73 32 Kapton 29 Kapton 6 single crystal (Bridgman) 150 350 T/K 1750 1500 μ,/ cm²V¹s⁻¹ 1250 p/10<sup>19</sup>cm<sup>3</sup> 500 250 350 100 300 100 T/K T/K

concentration increases. By considering the common defect structure model, it seems that the Te concentration becomes smaller if the films are thicker.

Increasing the amount of additional Te in the evaporant to 6 at%, results in a high mobility and small carrier concentration even in thick films.

 $Bi_2(Te_{0.9}Se_{0.1})_3$  films were doped with 0.04 at% J and are always n-type. Fig. 5 shows  $S^2\sigma$ ,  $\mu_H$  and the carrier concentration n in the same way as Fig. 4 does for p-type films. It was tried to optimize the thermoelectric properties by varying the contents of tellurium in the evaporant. As shown, the highest power factor was reached at 12 at% additional Te, which was the highest amount of Te within the experiments, whereas the highest mobility was reached at 8 at% Te.

There is still some more work to do to improve the carrier mobility in  $n-Bi_2(Te_{0.9}Se_{0.1})_3$  films and to study the influence of different dopant concentration.

Fig. 5: Properties of  $\mathrm{Bi}_{2}(\mathrm{Te}_{0.9}\mathrm{Se}_{0.1})_{3}$  films grown at  $T_s$ =200...230°C by flash evaporation So/ 103 WK 2m.1 Source material:  $\mathrm{Bi_2(Te_{0.9}Se_{0.1})_3}$  + add. Te +Te (at%) d (µm) substrat 0.59 Kapton 6 Kapton 8 1.60 Kapton 12 1.15 single crystal (Czochalski) 2 200 250 T/K 350 150 500 400 H, cm² V¹s.¹ n/10<sup>18</sup> cm<sup>-3</sup> 100 200 250 T/K 100 300 100 150